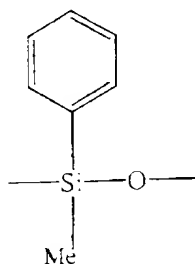


23. (new) A composition according to claim 1, wherein the poly(methylphenylsiloxane) comprises subunits of the formula



in an amount of greater than 90% of the total number of subunits in the poly(methylphenylsiloxane).

24. (new) A composition according to claim 23, wherein the poly(methylphenylsiloxane) further comprises dimethoxysiloxane subunits.

REMARKS

This is in response to the Office Action mailed February 21, 2003 for the above-captioned application. Reconsideration and further examination are respectfully requested.

The Examiner rejected claims 2-4 under 35 USC § 112, first paragraph. Claims 2-4 have been cancelled.

Claims 19-24 have been added. These amendments are supported on Page 7 of the application. Claims 1 and 5-24 are therefore pending. The Commissioner is authorized to charge the fee for additional claims to Deposit Account No. 07-0862.

The Examiner pointed to an error in antecedent basis in claim 15. Claim 15 has been amended to replace Rimar salt with potassium-perfluorobutane-sulfonate to provide proper dependence on claim 14. This is consistent with the statement equating these two names on Page 1, line 21 of the specification.

The Examiner rejected claims 1-18 under 35 USC § 112, second paragraph, stating that the term "salt based" is indefinite. The Examiner states: "it is not known which particular

property of which salt are the claimed composition based on." Applicants respectfully traverse this rejection.

The term "salt based" is an adjective which modifies the word flame retardant. As would be appreciated based on the common usage of the term, it indicates that the flame retardant in the composition is in the form of a "salt" as opposed to a non-ionizable charge neutral component. This is reflected in the statement on Page 7 of the application that "useful salt based flame retardants include alkali metal or alkaline earth metal salts of inorganic protonic acids as well as organic Brönsted acids." The Examiner statement of the rejection is therefore not understood. The particular chemical characteristic of the flame retardant is that it is a salt. The particular property of the salt that makes it relevant to the claims is that it acts as a flame retardant. The Examiner has not explained how a person skilled in the art would have difficulty understanding the scope of the claims when read in light of the specification. Accordingly, the Examiner has not met the burden associated with the presentation of a valid rejection under 35 USC § 112, second paragraph. *In re Cordova*, 10 U.S.P.Q. 2d 1949, 1952 (POBAI 1989).

The Examiner 1-18 under 35 USC § 102 as anticipated by any of four primary references, or as obvious over any of these references in combination with US Patent 6,454,969. Applicants respectfully submit that this rejection, in each case, reflects a misunderstanding of the scope of the invention on the part of the Examiner. Independent claim 1 in this application recites several components, one of which is poly(methylphenylsiloxane). In order for any of the four primary reference to anticipate this claim, they would need to disclose this specific siloxane, as it is defined on Page 6, line 23-Page 7, line 11 of the present specification. Such a disclosure is not found in the references cited.

In this regard, Applicants direct the Examiner's attention to MPEP § 2131.02, which states that:

When the compound is not specifically named, but instead it is necessary to select portions of teachings within a reference and combine them, e.g., select various substituents from a list of alternatives given for placement at specific sites on a generic chemical formula to arrive at a specific composition, anticipation can only be found if the classes of substituents are sufficiently limited or well delineated. *Ex parte A*, 17 USPQ2d 1716 (Bd. Pat. App. & Inter. 1990). If one of ordinary skill in the art is able to "at once envisage" the specific compound within the generic chemical formula, the compound is anticipated. One of ordinary skill in

the art must be able to draw the structural formula or write the name of each of the compounds included in the generic formula before any of the compounds can be "at once envisaged." One may look to the preferred embodiments to determine which compounds can be anticipated. In re Petering, 301 F.2d 676, 133 USPQ 275 (CCPA 1962).

With respect to Bialous et al. discloses siloxanes with a broad formula. If one makes specific selections from this formula, one can arrive at poly(methylphenylsiloxane). This however, is not anticipation because there is no teaching of the specific selections necessary and nothing in the reference which would guide a person skilled in the art to immediately envisage a poly(methylphenylsiloxane) as a species within the genus. Furthermore, the specific disclosure referred to by the Examiner is actually "linear bis(pentafluorophenyl) siloxane branched methylphenyl siloxane copolymer." The Examiner has not indicated how he in interpreting this language to fall within the scope of poly(methylphenylsiloxane) as required by the present claims.

With respect to Mark et al., the Examiner states that methylphenylsiloxane is disclosed at Col. 6 lines 50-51. Again, what is actually disclosed is "linear bis(pentafluorophenyl) siloxane branched methylphenyl siloxane copolymer," and the Examiner has not indicated how he in interpreting this language to fall within the scope of poly(methylphenylsiloxane) as required by the present claims.

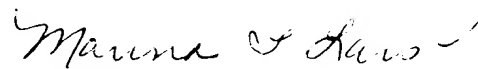
With respect to Umeda et al. the Examiner states that there is a siloxane disclosed in which R_4 may be methyl and/or phenyl. This is simply one choice of numerous combinations however, and there is no specific teaching in the reference of the specific combination of a poly(methylphenylsiloxane) or a reason to envisage such a composition given the nature of the specific.

Finally, with respect to the NEC abstract, this reference describes a polysiloxane that has as its principal characteristic the fact that it is branched. Applicants now enclose a translation of the complete Japanese application. As is apparent from the discussion of this substance on Page 10-11, there is no specific disclosure of a poly(methylphenylsiloxane). Thus, this reference does not anticipate the claimed invention.

In the alternative, the Examiner has asserted that it would have been obvious "to use the salts of Nishihara in the compositions of the primary references." Applicants do not understand the basis for the Examiner's argument. Replacing the salts of the references with the salts of Nishihara, as disclosed at Col. 15, line 29 et seq. would not overcome the deficiency of the primary references with respect to the siloxane components. If the Examiner intended to argue that replacing the siloxane would have been obvious, Applicants submits that this still does not support the rejection since even with the most careful selection, Nishihara does not disclose a poly(methylphenylsiloxane). Because of the oxygen atoms in formula 1, selection of methyl for R_1 and phenyl for R_2 (a combination which is not specifically taught or suggested by Nishihara) would result in poly(methoxyphenoxysiloxane). Thus, it is not clear how Nishihara would lead a person to modify the primary references to arrive at the invention as presently claimed.

For these reasons, this application is now considered to be in condition for allowance and such action is earnestly solicited.

Respectfully Submitted,



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MAY 07 2003
GROUP 1700

Japan Patent Office(JP) Patent application publication
Unexamined patent publication bulletin(A) JP2000-327897

43.Publication date 28.11.2000

51 Int. Cl. ⁷	Identification No.	FI
C08 L 69 00		C08 L 69 00
C08 K 5 42		C08 K 5 42
5 43		5 43
C08L 69 00		5 523

Request for Examination Yet not requested No. of claims 11 (Total pages 19)

21 Patent Application no. JP11-143840

22 Application Date 1999.05.24

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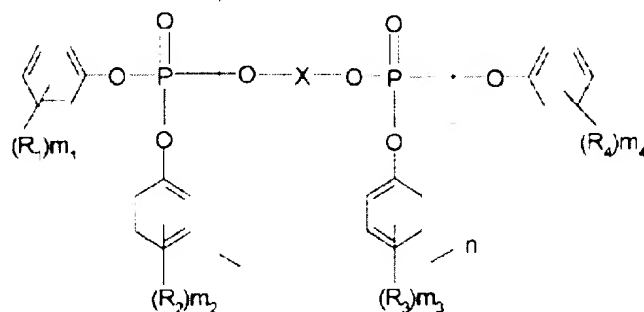
54 Title of the Invention Fire retardant polycarbonate resin composition

(57) Abstract

[Means for solving the problem]

The fire retardant polycarbonate resin composition is blended with 5 to 15 parts by weight phosphorous fire retardant (B) represented by formula 1, 0.05 to 8 parts by weight silicon compound (C) containing branched structures in the chain and aromatic group as aromatic substituent group, 0.05 to 5 parts by weight fibrous fluoro polymer (D), and 0.5 to 10 parts by weight elastomer (E), or less than 0.5 parts by weight of organic alkali metal salt (F) of aromatic sulfoimide, aromatic sulfonic acid, perfluoro alkane sulfonic acid, and aromatic sulfonic acid with respect to 100 parts by weight polycarbonate resin (A). Formula 1:

圖部、纖維狀樹脂の添加剤の一般式 (1) (2) (3) (4) (5)



(wherein, X is phenyl group, R₁ to R₄ is C₁₋₃ alkyl group, m₁ to m₄ is from 0 to 2, and n may be 1 to 5).

[Result]

The fire retardancy and the improvement in weak impact strength are achieved by the addition of phosphorous fire retardant, and can suitably used for an ultrathin molded articles.

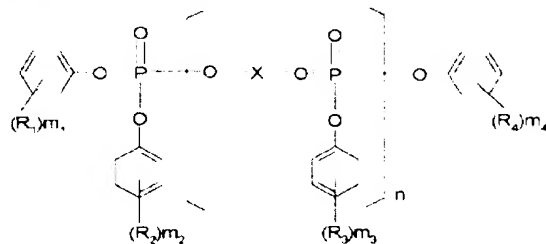
[Claim(s)]

[Claim 1] The fire retardant polycarbonate resin composition is characterized by blending with 5 to 15 parts by weight phosphorous fire retardant (B) represented by formula 1, 0.05 to 8 parts by weight silicon compound (C) containing branched structures in the chain and aromatic group as aromatic substituent group, 0.05 to 5 parts by weight fibrous fluoro polymer (D), and 0.5 to 10 parts by weight elastomer (E) with respect to 100 parts by weight of polycarbonate resin.

General formula 1:

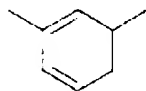
[Formula 1]

圖部、纖維狀樹脂の添加剤の一般式 (1) (2) (3) (4) (5)



wherein, X represents

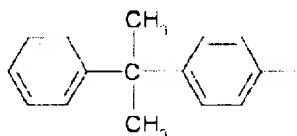
[Formula 2]



[Formula 3]



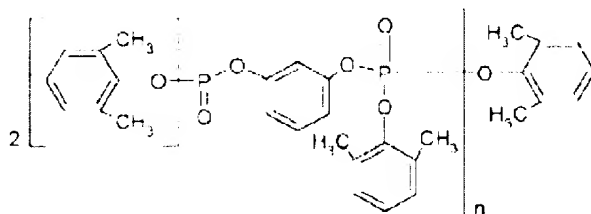
[Formula 4]



[Claim 2]

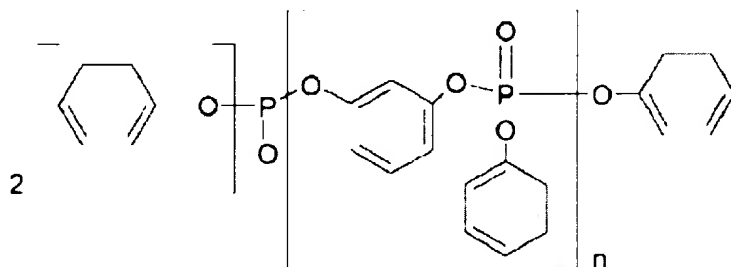
[Claim 3]

[Formula 5]



[Claim 4]

[Formula 6]



(wherein, n can have the value from 1 to 5).

[Claim 5]

The fire retardant polycarbonate resin composition mentioned in claim 1 or 2, wherein silicon compound (B) containing branched structures in the chain and aromatic group as aromatic substituent group contains atleast 20 mol% $\text{RSiO}_{1.5}$ unit (T unit), or $\text{SiO}_{2.0}$ unit (Q unit) with respect to total silicon unit ($\text{R}_{3.0}\text{SiO}_{2.5}$). (R represents organic substituent group).

[Claim 6]

The fire retardant polycarbonate resin composition mentioned in claim 5, wherein in silicon compound (B) containing branched structures in the chain and aromatic group as aromatic substituent group, aromatic group is atleast 20 mol%.

[Claim 7]

The fire retardant polycarbonate resin composition mentioned in claim 1 or 2, or claim 5 or 6, wherein in silicon compound (B) containing branched structures in the chain and aromatic group as aromatic substituent group, aromatic group is phenyl group, rest are methyl groups, terminal group is mixture of atleast one kind or four kinds selected from methyl, phenyl, hydroxyl, and alkoxy group.

[Claim 8]

The fire retardant polycarbonate resin composition mentioned in claim 1 or 2, wherein fibrous fluoro polymer (D) is polytetrafluoroethylene.

[Claim 9]

The fire retardant polycarbonate resin composition mentioned in claim 1 or 2, wherein elastomer (E) is atleast one kind selected from the core shell methyl methacrylate.styrene.butadiene rubber, core shell methyl methacrylate.styrene.acrylic ester rubber, core shell methyl methacrylate. glycidyl methacrylate. styrene.acrylic ester rubber, core shell methyl methacrylate. styrene.silicon.acrylic ester rubber, core shell methyl methacrylate. styrene.acrylonitrile.acrylic ester rubber, styrene.hydrogenated polyisoprene block copolymer.

[Claim 10]

The fire retardant polycarbonate resin composition mentioned in claim 1, wherein organic alkali metal salt (F) of aromatic sulfoimide, aromatic sulfonic acid, perfluoro alkane sulfonic acid, and aromatic sulfonic acid is less than 0.5 parts by weight with respect to 100 parts by weight of

polycarbonate resin (A).

[Claim 11]

The fire retardant polycarbonate resin composition mentioned in claim 10, wherein organic alkali metal salt (F) is at least one kind selected from potassium salt of N-(p-tolylsulfonyl)-p-toluene sulfoimide, potassium diphenylsulfone-3-sulfonate, potassium perfluoro butane sulfonate, and sodium p-toluene sulfonate.

Detailed description of the Invention

[0001]

[Industrial Field of Application]

The invention relates to the fire retardant polycarbonate resin composition. Specifically, it relates to fire retardant polycarbonate resin composition used for ultrathin molded articles having improved impact strength, mechanical properties, fluidity, fire retardancy without affecting the appearance of the molded article and not containing chloride and iodide compound of halogenated fire retardants.

[0002]

[Prior art]

Polycarbonate resin is widely used in electrical, electronic, and OA field as engineering plastic as it excels in transparency, impact strength, and electrical properties.

[0003]

In electrical, electronic, and OA field, the components requiring high fire retardancy (UL94V), impact strength, fluidity, heat resistance are quite large in number like battery case for lithium battery. Although, polycarbonate resin is plastic material having high fire retardancy with self-extinguishing properties, but in order to meet all the requirements in electrical, electronic, and OA field, high fire retardancy of UL94-V or 94V-1 is desirable. The needs of a market were not satisfied by polycarbonate resin independently.

[0004]

The method of blending phosphorous fire retardant, such as resorcin polyphosphate adequately is proposed in the past to improve the fire retardancy of polycarbonate resin.

[0005]

[Problem to be solved by the invention]

However, when resorcin polyphosphate is added adequately, it certainly improves the fire retardancy and fluidity of polycarbonate resin, but has a fatal flaw of weak impact strength and heat resistance.

[0006]

The impact resistance cannot be improved by blending with adequate amount of elastomer, and possesses a new problem of weak fire retardancy strength and heat resistance. The material having excellent balance of fire retardancy, impact strength, fluidity, and heat resistance, and satisfying the market need was not obtained.

[0007]

[Means for Solving the Problems]

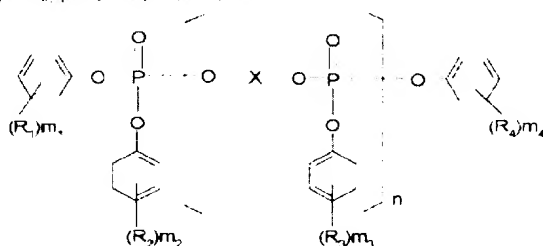
As a result of extensive research for the above-mentioned problem, silicon compound was used along with phosphorous fire retardant. Furthermore, addition of fibrous fluoro polymer and elastomer formed the resin composition with higher fire retardancy without affecting the impact strength, moldability, and fluidity, and the invention was completed.

[0008]

In other words, the resin composition is characterized by blending with 5 to 15 parts by weight phosphorous fire retardant (B) represented by formula 1, 0.05 to 8 parts by weight silicon compound (C) containing branched structures in the chain and aromatic group as aromatic substituent group, 0.05 to 5 parts by weight fibrous fluoro polymer (D), and 0.5 to 10 parts by weight elastomer (E) with respect to 100 parts by weight of polycarbonate resin.

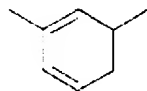
General formula 1:

[Formula 7]



wherein, X represents

[Formula 8]

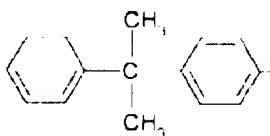


[Formula 9]



or,

{Formula 10}



R_1 to R_4 is $C_1 - 3$ alkyl group, m_1 to m_4 is from 0 to 2, and both can differ also. n may be 1 to 5).

[0009]

As one of the aspects of the invention, the fire retardant polycarbonate resin composition is characterized by blending with organic alkali metal salt (F) of aromatic sulfoimide, aromatic sulfonic acid, perfluoro alkane sulfonic acid, and aromatic sulfonic acid is less than 0.5 parts by weight with respect to 100 parts by weight of polycarbonate resin (A).

Hereinafter, the invention is explained briefly.

[0010]

Polycarbonate resin (A) used in the invention is a polymer obtained from phosgene method by reacting various dihydroxy diaryl compound and phosgene, or transesterification reaction of dihydroxy diaryl compound and carbonic acid ester, such as diphenyl carbonate. As an example, polycarbonate resin prepared from 2,2-bis(4-hydroxyphenyl)propane (bisphenol-A) is mentioned.

[0011]

As dihydroxy diaryl compound, other than bisphenol A, bis(hydroxyaryl) alkane series, such as bis(4-hydroxyphenyl)methane, 1,1-bis(4-hydroxyphenyl)ethane, 2,2-bis(4-hydroxyphenyl)butane, 2,2-bis(4-hydroxyphenyl)octane, bis(4-hydroxyphenyl) phenylmethane, 2,2-bis(4-hydroxyphenyl-3-methylphenyl)propane, 1,1-bis(4-hydroxy-3-tertiary butyl phenyl)propane, 2,2-bis(4-hydroxy-3-bromophenyl)propane, 2,2-bis(4-hydroxy-3,5-dibromophenyl)propane, 2,2-bis(4-hydroxy-3,5-dichlorophenyl)propane; bis(hydroxyaryl)cycloalkane series, such as 1,1-bis(4-hydroxyphenyl)cyclopentane, 1,1-bis(4-hydroxyphenyl)cyclohexane; dihydroxy diaryl ether series, such as 4,4'-dihydroxy diphenyl ether, 4,4'-dihydroxy-3,3'-dimethyl diphenyl ether; dihydroxy diaryl sulfide, such as 4,4'-dihydroxy diphenyl sulfide; dihydroxy diaryl sulfoxide, such as 4,4'-dihydroxy diphenyl sulfoxide, 4,4'-dihydroxy-3,3'-dimethyl diphenyl sulfoxide; dihydroxy diaryl sulfone, such as 4,4'-dihydroxy diphenyl sulfone, 4,4'-dihydroxy-3,3'-dimethyl diphenyl sulfone, are listed.

[0012]

These can be used independently, or in combination of at least two kinds, and halogen non-substituted are preferred from the viewpoint of prevention of discharge of gas containing halogen during combustion. Other than these, piperazine, dipiperidyl hydroquinone, resorcin, 4,4'-dihydroxy diphenyl can be used after mixing.

[0013]

In addition, mixture of dihydroxy diaryl compound and at least trivalent phenol compound can also be used.

[0014]

As trivalent phenol, phloroglucinol, 4,6-dimethyl-2,4,6-tri-(4-hydroxyphenyl)-heptane,

2,4,6-dimethyl-2,4,6-tri-(4-hydroxyphenyl)-heptane, 1,3,5-tri-(4-hydroxyphenyl)-benzol, 1,1,1-tri-(4-hydroxyphenyl)-ethane, and 2,2-bis-[4,4'-(4,4'-dihydroxy diphenyl)-cyclohexyl]-propane etc., are listed.

[0015]

The viscosity-average molecular weight of polycarbonate (A) is, generally, 10000 to 100000, preferably 15000 to 35000. During the preparation of polycarbonate resin, molecular weight adjuster, catalyst etc. can be added as per the requirement.

[0016]

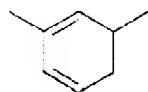
Phosphorous fire retardant (B) used in the invention is the compound represented by the given formula 1.

General formula 1:

[Formula 11]

wherein, X represents

[Formula 12]

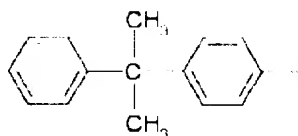


[Formula 13]



or,

[Formula 14]



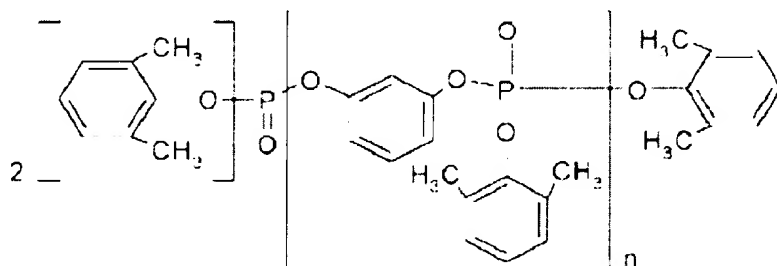
R_1 to R_4 is C_{1-3} alkyl group, m_1 to m_4 is from 0 to 2, and both can differ also. n may be 1 to 5).

[0017]

Specifically, the compounds represented by the given formulas 2 and 3 can be used suitably.

General formula 2:

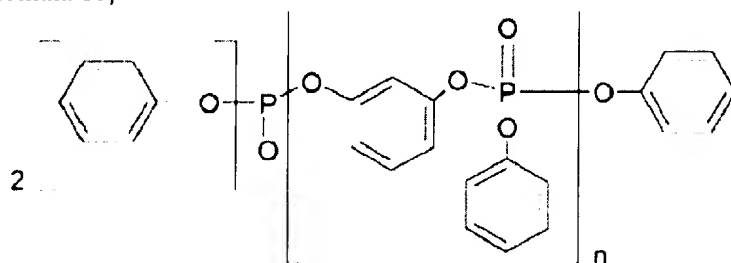
[Formula 15]



Wherein, n may have the value from 1 to 5.

General formula 3:

[Formula 16]



Wherein, n may have the value from 1 to 5.

[0018]

The blending amount of phosphorous fire retardant (B) is 5 to 15 parts by weight with respect to 100 parts by weight of polycarbonate resin (A). If the blend amount is less than 5 parts by weight, the fire retardancy effects are insufficient, and if exceeds 15 parts by weight, the impact strength is reduced considerably. The preferred range is 6 to 14 parts by weight, and most preferred is 8 to 12 parts by weight. In the above range, the balance of fire retardancy, moldability, impact strength, and heat resistance is substantially better. Moreover, monophosphoric acid ester compound can be used along with phosphorous fire retardant (B).

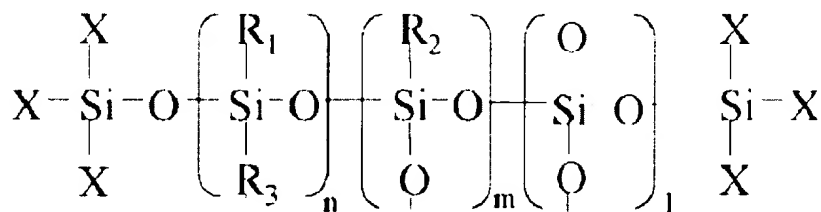
[0019]

Silicon compound (C) containing branched structures in the chain and aromatic group as aromatic substituent group is represented by the given formula 4.

[0020]

General formula 4:

[Formula 17]



Here, R_1 , R_2 , and R_3 are organic substituent groups of the chain, X is the terminal substituent group, n, m, and l represents mole number of the unit respectively.

[0021]

Silicon compound as fire retardant is formed by the given silicon unit (M unit, D unit, T unit, Q unit).

[0022]

1. M unit

Structure

Chemical formula

R

構造

R-Si-O-

化学式 $\text{R}_3\text{SiO}_{0.5}$

R

Wherein, R represents organic substituent group.

[0023]

2. D unit

Structure

Chemical formula

R

|

構造

-O-Si-O-

化学式 $\text{R}_2\text{SiO}_{1.5}$

|

R

Wherein, R represents organic substituent group.

[0024]

3. T unit

Structure

Chemical formula

R

|

構造

-O-Si-O-

化学式 $\text{R}_3\text{SiO}_{1.5}$

|

O

|

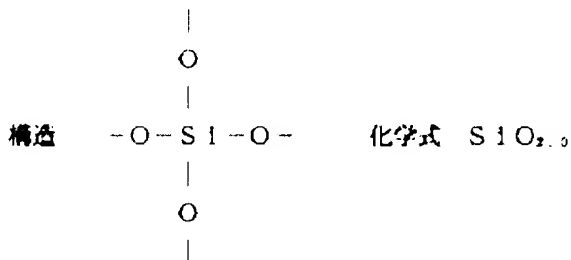
Wherein, R represents organic substituent group.

[0025]

4. Q unit

Structure

Chemical formula



[0026]

Amongst the all, the branched structures contain, specifically, T unit and/or Q unit. Silicon compound (C) used in the invention is characterized by containing T unit and/or Q unit as branched structures. It is preferred to contain atleast 20 mol% of the total silicon unit. If it is less than 20 mol%, the heat resistance and fire retardancy effects are affected considerably, and further the viscosity of silicon compound (C) falls down substantially that have adverse impact on kneading and moldability. The preferred range is 30 to 95 mol%. If it is atleast 30 mol%, the heat resistance of silicon compound (C) increases, and the fire retardancy of polycarbonate resin is also improved considerably. If it exceeds 95 mol%, the flexibility of the chain of silicon compound decreases it may be difficult to bring about condensation of aromatic environment during combustion, and may be difficult to develop remarkable fire retardancy.

[0027]

In addition, aromatic group of organic substituent group contained in silicon compound (C) is preferred to be 20 mol%. If it less than the range, it is difficult to carry out the condensation of aromatic groups during combustion, and also fire retardancy effects are affected. The preferred range is 40 to 95 mol%. If it is atleast 40 mol%, along with the possibility of the condensation of aromatic groups, the dispersibility of silicon compound (C) improves considerably, and better fire retardancy effects are discovered. If exceeds 95 mol%, the condensation becomes difficult because of steric hindrance of aromatic groups, and becomes difficult to develop remarkable fire retardancy.

[0028]

The aromatic group may be phenyl, biphenyl, naphthalene, or their derivatives, but from the viewpoint of safe condition of silicon compound (C), specifically phenyl is preferred. Moreover, as organic substituent of silicon compound (C), methyl group is preferred to be attached with the side chain or main chain as organic group, other than aromatic group. In addition, the terminal group is the mixture of one kind or from two kinds to four kinds selected from methyl, phenyl, hydroxyl, alkoxy (specifically, methoxy) groups. When the terminal group is from the above mentioned groups,

the reactivity is low, the gelatinization of silicon compound (C) is not possible during kneading, and because of this reason silicon compound (c) is uniformly dispersed in polycarbonate resin (A). As a result, it is possible to have better fire retardancy effects, and moldability is also improved. The most preferred is methyl group. In this case, because of extremely low reactivity, dispersibility becomes better, and fire retardancy can be improved.

[0029]

The preferred average molecular weight of silicon compound (C) is 5000 to 500000. If it is less than 5000, the heat resistance and fire retardancy effects are reduced. Moreover, melt viscosity falls down considerably, silicon compound is leached out on the surface of the molded article of polycarbonate resin (A) during molding, and moldability is affected. If it exceeds 500000, the uniform dispersibility in polycarbonate resin (A) is affected by increase in melt viscosity, and fire retardancy and moldability are affected. The preferred range is 10000 to 270000. In the above mentioned range, the uniform dispersibility of silicon compound (C) is possible in polycarbonate (A) as the melt viscosity of silicon compound (C) is ideal, and fire retardancy and moldability can be improved as there is no excess leaching on the surface.

[0030]

The blend amount of silicon compound (C) is 0.05 to 8 parts by weight to 100 parts by weight of polycarbonate resin (A). In this case, not only the fire retardancy but impact strength is also improved. If the blend amount is less than 0.05 parts by weight, the fire retardancy effects are inadequate. If it exceeds 8 parts by weight, layer exfoliation is observed on the surface of the molded article with inferior appearance. The preferred range is 0.1 to 5 parts by weight, and most preferred range is 0.1 to 2 parts by weight. In this range, the balance of fire retardancy, moldability, and impact strength is substantially better.

[0031]

The fibrous fluoro polymer (D) used in the invention can be formed by the fibre structures in polycarbonate resin (A). Polycarbonate formed from polytetrafluoroethylene, tetrafluoroethylene copolymer (e.g., tetrafluoroethylene hexafluoropropylene copolymer, etc.), various partial fluoro polymer mentioned in US patent 4379910, fluorinated phenol are also listed. The dripping prevention effect is observed when phosphorous fire retardant (B), silicon compound (C), and elastomer (E) are used together.

[0032]

The blend amount of fibrous fluoro polymer (D) is 0.05 to 5 parts by weight to 100 parts by weight of polycarbonate resin (A). When the blend amount is less than 0.05 parts by weight, the dripping prevention effect is mild during combustion, and if exceeds 5 parts by weight, the pelletizing will become difficult, and interfere with safe production. The preferred range is 0.05 to 1 part by weight.

and most preferred is 0.1 to 0.5 parts by weight. In this range, the balance of fire retardancy, moldability, impact strength, and pelletization is substantially better.

[0033]

The examples of elastomer (E) used in the invention are core shell methyl methacrylate.styrene.butadiene rubber, core shell methyl methacrylate.styrene.acrylic ester rubber, core shell methyl methacrylate, glycidyl methacrylate, styrene.acrylic ester rubber, core shell methyl methacrylate, styrene.silicon.acrylic ester rubber, core shell methyl methacrylate.styrene.acrylonitrile.acrylic ester rubber, ethylene.propylene rubber, ethylene.propylene.diene.methylene rubber, polyester elastomer, polyester.polyether copolymer, polyacrylic ester, polyisoprene, hydrogenated polyisoprene, styrene.hydrogenated polyisoprene block copolymer, polyamide elastomer, ethylene. vinyl acetate copolymer, ethylene. vinyl acetate.glycidyl methacrylate copolymer, ethylene.acrylic ester copolymer, ethylene.acrylic ester rubber.maleic anhydride copolymer, ethylene.acrylic ester, glycidyl methacrylate copolymer, hydrogenated styrene.ethylene.butylene.styrene block copolymer, ethylene.polyacrylic ester, carbon monoxide copolymer. These can be used independently or in combinations.

[0034]

The suitable elastomers are core shell methyl methacrylate.styrene.butadiene rubber, core shell methyl methacrylate.styrene.acrylic ester rubber, core shell methyl methacrylate, glycidyl methacrylate, styrene.acrylic ester rubber, core shell methyl methacrylate, styrene.silicon.acrylic ester rubber, core shell methyl methacrylate.styrene.acrylonitrile.acrylic ester rubber, and styrene.hydrogenated polyisoprene block copolymer.

[0035]

The blend amount of elastomer (E) is 0.5 to 10 parts by weight to 100 parts by weight of polycarbonate resin (A). If the blend amount is less than 0.5 parts by weight, the improved effects of impact strength are not obtained, and if exceeds 10 parts by weight, the impact strength is further improved but fire retardancy decreases considerably, and hence not required. The preferred range is 1 to 8 parts by weight, and most preferred range is 2 to 6 parts by weight. In this range, the balance of fire retardancy, moldability, and impact strength is substantially better.

[0036]

Organic alkali metal salt (F) is atleast one kind or other organic metal salt selected from the alkali metal salts of aromatic sulfoimide, aromatic sulfonic acid, perfluoro alkane sulfonic acid, and aromatic sulfonic acid. Potassium salt of N-(p-tolylsulfonyl)-p- toluene sulfoimide, potassium diphenylsulfone-3-sulfonate, potassium perfluoro butane sulfonate, and sodium p-toluene sulfonate can be used suitably.

[0037]

The blend amount of organic alkali metal salt (F) is less than 0.5 parts by weight with respect to 100 parts by weight of polycarbonate. If it exceeds 0.5 parts by weight, the heat stability during injection molding is inferior.

[0038]

Polycarbonate resin (A) can be blended with various heat stabilizer, antioxidants, pigments, such as titanium oxide or carbon black, fluorescent whitener, filler, mold lubricant, softener, antistatic agent, UV absorbent, other polymer, as long they does not have any adverse effect on the invention.

[0039]

Phosphite and phosphonite compounds as heat stabilizer, and phenol compounds as antioxidants can also be blended.

[0040]

The pigment titanium oxide used in the invention can be the product prepared by chlorination or sulfonation process, and these crystalline structures can be rutile, anatase structures. Moreover, titanium oxide can also use the products that are surface treated by silicon compound, e.g. various silane coupling agent or polymethyl hydrogen siloxane.

[0041]

Glass fibre, glass beads, glass flakes, carbon fibre, talc granules, clay granules, mica, potassium titanate whisker, wollastonite granules, silica granules, etc. can be used as fillers.

[0042]

The other polymer used in the invention is polyester, such as polyethylene terephthalate, polybutylene terephthalate; styrene polymer, such as polystyrene, acrylonitrile-styrene copolymer; polypropylene, other polycarbonate resin, and polymers manufactured by alloying.

[0043]

There is no restriction for the mixing method of various components of fire retardant polycarbonate resin composition, e.g. fusion and kneading can be performed by extruder or other mixers, such as tumbler, ribbon blender etc..

[0044]

The molding methods used for the fire retardant polycarbonate resin composition are injection molding, ejection and compression molding etc..

[0045]

[Embodiment]

Hereinafter, the invention is explained with the help of embodiments, but the invention is not restricted to these embodiments only. Moreover, [parts] is based on weight standards.

[0046]

[Embodiment 1 to 39 and Comparative Example 1 to 24]

Various mixed compounds were mixed and kneaded by the amount shown in table 2 to 8 with

respect to 100 parts of polycarbonate resin, prepared from bisphenol-A, using 37 mm diameter biaxial extruder (Kobe Steel, Ltd. KTX-37), and various pellets were obtained.

[0047]

The details of each material used in the invention are as follows:

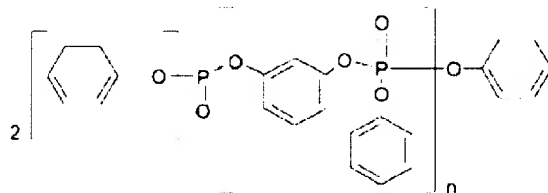
1. Polycarbonate resin (A): Sumitomo Dow Chemicals product, Calibre 200-10 (viscosity average molecular weight 22,4000)

[0048]

2. Phosphorous fire retardant (B):

Asahi Denka Chemicals Ltd., Adeka Stab PFR (hereinafter, abbreviated to B-1)

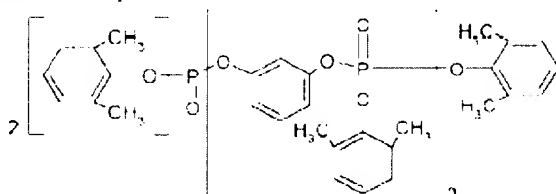
[Formula 18]



wherein, n varies from 1 to 5.

Adeka Stab FP500 (hereinafter, abbreviated to B-2).

[Formula 19]



wherein, n varies from 1 to 5.

[0049]

3. Silicon compound (C): Silicon compound (C) was prepared by general method. In other words, corresponding to molecular weight of silicon compound components and proportion of M unit, D unit, T unit, and Q unit that forms the silicon compound, the appropriate amount of diorgano dichloro silane, monoorgano trichloro silane, and tetrachloro silane, or their partially hydrolyzed condensed product were dissolved in organic solvent. It was hydrolyzed after adding water, condensed silicon compounds were molded partially, and polymerization reaction was completed after adding triorgano chlorosilane. The solvent was separated by distillation. The structural properties of 19 kinds silicon compound that were manufactured by the above mentioned method, is shown in Table 1.

[0050]

[Table 1]

Silicon	D/T/Q ratio of main chain structure (mol ratio)	Ratio of phenyl group* in all organic substituent group (mol%)	Ratio and structure of terminal group (mol ratio)	Molecular weight (weight average)**
A	0.7/9.3/0	60	methyl group	12,000
B	2/8/0	60	methyl group	7,000
C	2/8/0	60	methyl group	12,000
D	2/8/0	60	methyl group	70,000
E	2/8/0	60	methyl group	250,000
F	2/8/0	60	methyl group	300,000
G	2/8/0	60	hydroxyl group	250,000
H	5/5/0	40	methyl group	80,000
I	6.5/3.5/0	50	methyl group	50,000
J	6.5/1.5/2	50	methyl group	50,000
K	6.5/3.5/0	50	methyl group methoxy group = 1/1	50,000
L	7.5/2.5/0	50	methyl group	50,000
M	9/1/0	50	methyl group	50,000
N	10/0/0	50	methyl group	50,000
O	2/8/0	90	phenyl group	70,000
P	2/8/0	45	methyl group	70,000
Q	2/8/0	25	methyl group	70,000
R	2/8/0	10	methyl group	70,000
S	2/8/0	0	methyl group	70,000

*: phenyl group is contained in T unit in silicon compound containing T unit, and rest is contained in D unit. When phenyl group is attached to D unit, the first group attached has the priority, and when phenyl group is left, two groups are attached. Leaving the terminal group, all are methyl group other than phenyl group.

** : 2 digit significant number is weight average molecular weight.

[0051]

4. Fibrous fluoro polymer (D):

Polytetrafluoroethylene (Daikin product, TFE FA-500) (hereinafter, abbreviated to PTFE).

[0052]

5. Elastomer (E): Kureha Chemical Industry Co., Ltd product, Paraloid EXL 2602 (abbreviated to E-1).

(Core shell methyl methacrylate.styrene.butadiene rubber): Kureha Chemical Industry Co., Ltd product, Paraloid EXL 2314 (abbreviated to E-2).

(Core shell methyl methacrylate.glycidyl methacrylate.styrene.acrylic ester rubber): Takeda Chemical Industries, Ltd. product, Staphyloid AC1013 (abbreviated to E-3).

(Coreshell methyl methacrylate.styrene.acrylonitrile.acrylic ester rubber): Mitsubishi Rayon product, Metablen S2001 (abbreviated to E-4).

(Coershell methyl methacrylate.styrene.silicon.acrylic ester rubber)

[0053]

6. Organic alkali metallic salt (F):

Potassium salt of N-(p- tolylsulfonyl)-p-toluenesulfoimide (abbreviated to F-1).

Potassium diphenyl sulfone-3-sulfonate (abbreviated to F-2).

Potassium perfluoro butane sulfonate (abbreviated to F-3).

Sodium p-toluene sulfonate (abbreviated to F-4).

[0054]

After drying the obtained pellets at 90 to 125°C for 8 hrs, the test sample (127 X 12.7 X 0.75 mm) for flame retardancy evaluation was prepared using injection molding machine (Japan Steel Works, Ltd. product, J100-E-C5) at melting temperature of 270 to 300°C.

[0055]

The test sample was left for 48 hrs in temperature-controlled room at 23°C and 50% humidity. The flame retardancy evaluation was carried out in conformity to Underwriter's Laboratories UL94 test. The UL94 test is a test in which a standard specimen clamped in the vertical position is exposed to a burner flame. The flame is applied to the sample for ten seconds after which it is removed and an after flame time and dripping is used to evaluate flammability. They are classified as follows:

	V-0	V-1	V-2
After flame time of each sample	less than 10 sec	less than 30 sec	less than 30 sec
Total flame time of 5 samples	less than 50 sec	less than 250 sec	less than 250 sec
Cotton wool ignited by dripping	No	No	Yes

Afterflame time is the length of time for which a material continues to flame, under the specified test conditions, after the ignition source has been removed. Ignition of cotton wool is determined by

whether any specimen that drip flaming particles ignites the dry absorbent cotton located 300 mm below the test specimen or not. The results are shown in table 2 to 9.

[0056]

The injection molding was carried out using the obtained pellets, and test sample (3.2 X 12.7 X 63.5 mm) was prepared for impact strength evaluation. The impact strength was determined in conformity with ASTM D-256 at 23°C with notch. The numeric value atleast 35 was accepted. In addition, the appearance of the molded article was visually observed before the impact strength evaluation, and the surface exfoliation or depression was checked. Further, the resin was extruder in the form of strands inside the extruder, and the conditions during the preparation of pellets were visually observed. The results are shown in table 2 to 9.

[0057]

In table 2 to 9:

-The numerical value of phosphorous fire retardant, silicon compound, PTFE, elastomer, and organic metallic slats represents the amount with respect to 100 parts of polycarbonate resin.

-()value represents the total afterflame time (sec) in flammability evaluation results. []value represents the ignition of cotton wool by dripping. Flammability test UL94 was passed.

-Moldability was evaluated on the basis of surface exfoliation or surface depression of the test sample. O and Δ levels were passed.

O: Was not discovered

Δ: Was observed in 1 to 2 samples out of 5 samples.

X: Was observed in 3 samples out of 5 samples.

-Pelletizing was visually determined by the conditions during the preparation of pellets, when the resin was extruder in the form of strands inside the extruder. O and Δ levels were passed.

O: easy pelletization

Δ: slight difficulty in pelletization

X: pelletization was not possible

[0058]

[Table 2]

		Embodiment								
		1	2	3	4	5	6	7	8	9
Phosphorous	fire	8	8	8	8	8	8	8	8	8
retardant B-1										
silicon a		2	-	-	-	-	-	-	-	-
silicon b		-	2	-	-	-	-	-	-	-
silicon c		-	-	2	-	-	-	-	-	-
silicon d		-	-	-	2	-	-	-	-	-
silicon e		-	-	-	-	2	-	-	-	-
silicon f		-	-	-	-	-	2	-	-	-
silicon g		-	-	-	-	-	-	2	-	-
silicon h		-	-	-	-	-	-	-	2	-
silicon i		-	-	-	-	-	-	-	-	2
PTFE		0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5
Elastomer E-1		3	3	3	3	3	3	3	3	3
Flammability	0.8mm	V-0	V-0	V-0	V-0	V-0	V-0	V-0	V-0	V-0
UL-94 thickness		(27)	(28)	(22)	(18)	(18)	(29)	(28)	(22)	(25)
Notch strength (kg.cm/cm)	impact	63	62	64	64	65	63	64	63	62
Moldability		O		O	O	O		O	O	O
Pelletization		O	O	O	O	O	O	O	O	O

[0059]

[Table 3]

		Embodiment							
		10	11	12	13	14	15	16	17
Phosphorous retardant B-1 silicon j silicon l silicon m Silicon n silicon o silicon p silicon q silicon r	fire	8	8	8	8	8	8	8	8
		2	-	-	-	-	-	-	-
		-	2	-	-	-	-	-	-
		-	-	2	-	-	-	-	-
		-	-	-	2	-	-	-	-
		-	-	-	-	2	-	-	-
		-	-	-	-	-	2	-	-
		-	-	-	-	-	-	2	-
		-	-	-	-	-	-	-	2
PTFE		0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5
Elastomer E-1		3	3	3	3	3	3	3	3
Flammability UL-94 0.8mm thickness		V-0	V-0	V-0	V-0	V-0	V-0	V-0	V-0
		(25)	(30)	(35)	(39)	(28)	(23)	(33)	(37)
Notch strength (kg.cm/cm)	impact	63	63	61	59	66	64	62	56
Moldability		O	O	O		O	O	O	
Pelletization		O	O	O	O	O	O	O	O

[0060]

[Table 4]

		Embodiment							
		18	19	20	21	22	23	24	25
Phosphorous retardant B-1	fire	6	8	8	8	12	-	-	-
Phosphorous retardant B-2	fire	-	-	-	-	-	8	10	14
silicon d		2	0.1	4	7	7	2	0.1	7
PTFE		0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5
Elastomer E-1		3	3	3	3	3	3	3	3
Flammability U1-94		V-0	V-0	V-0	V-0	V-0	V-0	V-0	V-0
0.8mm thickness		(39)	(23)	(19)	(29)	(24)	(21)	(19)	(24)
Notch strength (kg.cm/cm)	impact	67	58	66	68	63	65	55	56
Moldability		O	O	O	O	O	O	O	O
Pelletization		O	O	O	O	O	O	O	O

[0061]

[Table 5]

		Embodiment								
		26	27	28	29	30	31	32	33	34
Phosphorous retardant B-1	fire	8	8	8	-	-	-	8	8	8
Phosphorous retardant B-2	fire	-	-	-	10	12	14	-	-	-
silicon e		2	2	2	2	2	2	2	2	2
PTFE		0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5
Elastomer E-1		3	3	3	1	5	8	-	-	-
Elastomer E-2		-	-	-	-	-	-	3	-	-
Elastomer E-3		-	-	-	-	-	-	-	3	-
Elastomer E-4		-	-	-	-	-	-	-	-	3
Flammability U1-94	0.8mm thickness	V-0	V-0	V-0	V-0	V-0	V-0	V-0	V-0	V-0
		(20)	(17)	(18)	(15)	(19)	(30)	(17)	(18)	(19)
Notch strength (kg.cm/cm)	impact	65	64	63	54	62	59	60	55	65
Moldability		O	O	O	O	O	O	O	O	O
Pelletization		O	O		O	O	O	O	O	O

[0062]

[Table 6]

	Embodiment				
	35	36	37	38	39
Phosphorous fire retardant B-1	8	8	8	8	8
silicon d	2	2	2	2	2
PTFE	0.5	0.5	0.5	0.5	0.5
Elastomer E-1	3	3	3	3	5
Organic metallic salt F-1	0.1	-	-	-	-
Organic metallic salt F-2	-	0.02	0.4	-	-
Organic metallic salt F-3	-	0.02	-	0.05	-
Organic metallic salt F-4	-	-	-	-	0.02
Flammability UL-94 0.8mm thickness	V-0 (13)	V-0 (47)	V-0 (15)	V-0 (13)	V-0 (14)
Notch impact strength (kg.cm/cm)	63	64	62	63	64
Moldability	O	O	O	O	O
Pelletization	O	O	O	O	O

[0063]

[Table 7]

	Comparative Example							
	1	2	3	4	5	6	7	8
Phosphorous fire retardant B-1	8	8	-	-	4	4	6	6
silicon n	2	-	-	-	-	-	-	-
silicon s	-	2	-	-	-	-	-	-
silicon d	-	-	2	7	-	2	-	0.02
PTFE	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5
Elastomer E-1	3	3	3	3	3	3	3	3
Flammability UL-94 0.8mm thickness	V-1 (52)	V-1 (53)	NR (196)	NR (220)	NR (153)	V-1 (102)	V-1 (71)	V-0 (62)
Notch impact strength (kg.cm/cm)	24	23	71	72	66	68	60	61
Moldability	X	X	O	O	O	O	O	O
Pelletization	O	O	O	O	O	O	O	O

[0064]

[Table 8]

Comparative Example							
	9	10	11	12	13	14	15
Phosphorous fire retardant B-1	8	8	8	16	16	16	16
silicon d	-	0.02	10	-	2	7	10
PTFE	0.5	0.5	0.5	0.5	0.5	0.5	0.5
Elastomer E-1	3	3	3	3	3	3	3
Flammability UL-94 0.8mm thickness	V-1 (45)	V-0 (38)	V-1 (47)	V-0 (16)	V-0 (15)	V-0 (22)	V-0 (29)
Notch strength impact (kg.cm/cm)	26	30	66	9	10	12	15
Moldability	O	O	X	O	O	O	X
Pelletization	O	O	O	O	O	O	O

[0065]

[Table 9]

Comparative Example									
	16	17	18	19	20	21	22	23	24
Phosphorous fire retardant B-1	8	8	8	-	-	-	-	-	-
Phosphorous fire retardant B-2	-	-	-	10	10	10	14	14	17
PTFE	-	0.03	7	0.5	0.5	0.5	0.5	0.5	0.5
Elastomer E-1	3	3	3	-	0.3	12	12	12	12
Flammability UL-94 0.8mm thickness	V-2 [12]	V-2 [6]	- -	V-0 (14)	V-0 (15)	NR (99)	V-1 (58)	V-1 (61)	V-1 (70)
Notch strength impact (kg.cm/cm)	68	68	-	18	22	69	62	64	26
Moldability	0	0	-	O	O	O	O	O	X
Pelletization	0	0	X	O	O	O	O	O	O

[0066]

Polycarbonate resin having phosphorous fire retardant (B) 5 to 15 parts, silicon compound having branched structures in chain and aromatic group (C) 0.05 to 8 parts, fibrous fluoro polymer (D) 0.05 to 5 parts, and elastomer (E) 0.5 to 10 parts as embodiments 1 to 34 has outstanding fire retardancy and impact strength as compared to polycarbonate resin not having phosphorous fire retardant (B), silicon compound (C), fibrous fluoro polymer (D), and elastomer (E)(comparative example 3 to 5, 7, 9, 12, 16, 19), or polycarbonate resin having structures of silicon compound other than the invention(comparative example 1, 2). Furthermore, the problem of weak impact strength of polycarbonate resin during the addition of phosphorous fire retardant (comparative example 10, 12 to 15, 19, 24) has been solved considerably in the embodiments.

[0067]

In order to combine fire retardancy and impact strength, it is essential to have phosphorous fire retardant (B) and silicon compound (C) as 5 to 15 parts and 0.05 to 8 parts respectively as in

embodiment 18 to 25 and comparative example 5 to 15. After comparing embodiment 18, 19 and comparative example 5 to 10, it is clear that if the amount of phosphorous fire retardant (B) and silicon compound (C) is less than 5 parts and 0.05 parts respectively, fire retardancy will be weak (comparative example 5). If amount of silicon compound (C) is in the specified range, but amount of phosphorous fire retardant (B) is less than 5 parts (comparative example 3,4,6), the fire retardancy would be low. If amount of phosphorous fire retardant (B) is in the specified range, but amount of silicon compound (C) is less than 0.05 parts (comparative example 7 to 10), the fire retardancy or impact strength would be low. In addition, if the amount of phosphorous fire retardant (B) is in the specified range, but amount of silicon compound (C) exceeds 8 parts (comparative example 11), would have inferior moldability. On the other hand, if the amount of phosphorous fire retardant (B) exceeds 15 parts (comparative example 12 to 14), the impact strength would be considerably low. Moreover, if the amount of phosphorous fire retardant (B) and silicon compound (C) exceeds 15 parts and 8 parts respectively (comparative example 15), not only impact strength but moldability is also affected.

[0068]

The fibrous fluoro polymer (D) is an indispensable additive agent to improve fire retardancy, and dripping prevention effects of polycarbonate resin during combustion. The amount of fibrous fluoro polymer (D) was 0.05 to 5 parts in the embodiments 26 to 28 and comparative examples 16 to 18. If the amount was less than 0.05 parts (comparative example 16, 17), dripping was observed during the combustion, and fire retardancy was low. Further, when the composition having 7 parts of fibrous fluoro polymer (D) was prepared, the pelletizing was extremely difficult, and pelletization was not possible.

[0069]

In order to improve the low impact strength by adding phosphorous fire retardant (B), only silicon compound (C) was not sufficient, but it was necessary to add 0.5 to 10 parts of elastomer (E). After comparing embodiment 29 to 31 and comparative example 19 to 24, it was clear that if the amount of elastomer (E) was less than 0.5 parts (comparative example 19, 20), the fire retardancy was better, but the impact strength was considerably low. If exceeds 10 parts (comparative example 21, 22, 23), the impact strength was better, but the fire retardancy was considerably low. Moreover, when phosphorous fire retardant (B) and silicon compound (C) was added exceeding the specified range (comparative example 24), not only fire retardancy impact strength but impact strength was also low. The composition with better fire retardancy and impact strength was obtained by changing the elastomer (E) type as shown in embodiment 4, 32 to 34.

[0070]

As the structures of silicon compound (C), if the chain contains branched units, such as unit of formula $\text{RSiO}_{1.5}$ (T unit), or unit of formula $\text{SiO}_{2.0}$ (Q unit) as embodiments 1 to 17, the fire

retardancy, moldability, and the impact strength is improved considerably as compared to silicon that does not contain the units (comparative example 1). Specifically, if it is contained at least 20 mol% with respect to total siloxane unit ($R_{3.0}SiO_{2.0.5}$) (embodiment 9 to 12), the fire retardancy is substantially better. After comparing embodiment 9 and comparative example 12, it is clear that if the branched unit is at least 30 mol% (embodiment 9), the fire retardancy is further improved. If the branched units exceeds 95 mol%, it is difficult to get the remarkable fire retardancy effects. It is preferred to have $RSiO_{1.5}$ (T unit), or unit of formula $SiO_{2.0}$ (Q unit) at least 20 mol% with respect to total siloxane unit ($R_{3.0}SiO_{2.0.5}$), preferably at least 30 mol% and less than 95 mol% from the viewpoint of fire retardancy, moldability, and impact strength. Further, if it contains $SiO_{2.0}$ (Q unit) as embodiment 10, the fire retardancy results would be same as the substance containing only $RSiO_{1.5}$ (T unit).

[0071]

In addition, if the composition contains aromatic group (phenyl) as organic substituent group in silicon compound (C) (embodiment 14 to 17), the fire retardancy, moldability, and impact strength is improved as compared to the composition not containing the organic substituent group (comparative example 2). Specifically, if it is contained at least 20 mol% (embodiment 14 to 16), the fire retardancy, moldability, and impact strength is substantially better. If it is 40 mol% (embodiment 14, 15), the fire retardancy is improved considerably. If aromatic group (phenyl) as organic substituent group exceeds 95 mol%, it is difficult to get the remarkable fire retardancy effects. It is preferred to have aromatic group (phenyl) as organic substituent group in silicon compound (C) at least 20 mol%, preferably at least 40 mol% and less than 95 mol% from the viewpoint of fire retardancy, moldability, and impact strength.

[0072]

When the terminal group of silicon compound (C) is methyl group (embodiment 1 to 6, 8 to 10, 15 to 17), phenyl group (embodiment 14), hydroxyl group (embodiment 7), alkoxy group: methoxy) (embodiment 11), the polycarbonate resin shows remarkable fire retardancy, moldability, and impact strength. Comparison of embodiment 5 and 7 shows that methyl group instead of hydroxy group, or methyl group instead of alkoxy group in embodiment 9 and 11, has better fire retardancy. Methyl group is preferred over phenyl group for better fire retardancy. When the terminal group is epoxy group (-glycidoxypropyl group) or vinyl group, because of high reactivity, the gelatinization of silicon compound (C) is possible during kneading, and the moldability of polycarbonate resin is affected. As a result, the fire retardancy effects and impact strength are inadequate because of low dispersibility in polycarbonate resin. Therefore, the terminal group is preferred to be methyl group.

[0073]

The preferred molecular weight of silicon compound is 5000 to 500000, preferably 10000 to

270000 as in embodiment 2, 3, 5, 6 from the viewpoint of moldability and fire retardancy.

[0074]

It was found that if organic alkali metallic salt (F) is added less than 0.5 parts, the fire retardancy is improved (embodiment 35 to 39).

[0075]

[Result of the invention]

The impact strength of fire retardant polycarbonate resin composition can be improved considerably after the addition of phosphorous fire retardant, and the fire retardancy and impact strength is also achievable. It is possible to offer a resin composition ideal for ultra thin molded articles.